Highly enhanced thermopower in two-dimensional electron systems at milliKelvin temperatures

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We report experimental observation of an unexpectedly large thermopower in mesoscopic two-dimensional (2D) electron systems on GaAs/AlGaAs heterostructures at sub-Kelvin temperatures and zero magnetic field. Unlike conventional non-magnetic high-mobility 2D systems, the thermopower in our devices increases with decreasing temperature below 0.3 K, reaching values in excess of 100 μV/K, thus exceeding the free electron estimate by more than two orders of magnitude. With support from a parallel independent study of the local density of states, we suggest such a phenomenon to be linked to intrinsic localized states and many-body spin correlations in the system.

The diffusion thermopower (TP), S_d, in a solid depends on the variation of scattering time or the density-of-states (DOS) in the vicinity of the Fermi energy (E_F), and expressed by the Mott relation [1, 2]:

$$S_d = \lim_{\Delta T \to 0} \frac{V_{th}}{\Delta T} = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{d \ln \sigma(E)}{dE}\Big|_{E=E_F} \quad (1)$$

where V_{th} is the thermovoltage at temperature T, and $\sigma(E)$ is the energy-dependent conductivity. Consequently, TP is directly sensitive to proximity to band edge or gaps in DOS in strongly localized systems. This has been exploited in investigating the localization transition in a disordered 2DES [3], where the 2DES goes from a purely metallic state to a highly localized one. Similar TP measurements have been used to explore quantum insulating ground states of a bulk 2D electron gas in high magnetic fields [4]. On the other hand, being a measure of entropy per carrier, the third law of thermodynamics requires TP $\to 0$ at $T \to 0$ in delocalized or metallic systems, although the absolute magnitude of TP depends critically on the energy-dependence of scattering mechanism of the conduction electrons. This has direct implications for a large classs of highly correlated 3D systems, [5, 6, 7, 8, 9, 10], such as the dilute magnetic alloys, heavy Fermions or Kondo lattice compounds. Here, the conduction electrons undergo spin-flip scattering at the localized $d-$ or $f-$ sites, which may lead to very large TP, exceeding several hundred μV/K [11, 12]. In this study, we focus on nonmagnetic delocalized high-mobility 2D electron systems (2DES) at GaAs/AlGaAs interface, which are not expected to contain localized spins, and indeed, earlier measurements of zero magnetic field diffusion TP in macroscopic 2DES yielded TP $\lesssim 0.4$ μV/K even at T as large as ≈ 3 K [13].

Recently though, several experiments on nonequilibrium transport in quasi-one dimensional quantum point contacts (QPC) [14, 15] and unconfined mesoscopic 2DES [16, 17, 18] suggest the possibility of localized spins in these III-V semiconductor-based low-dimensional systems. While the microscopic origin of the localized spins in such materials remains unclear, measurements in QPCs have indeed revealed additional contribution to the diffusion thermopower near the so-called '0.7 state' that is not entirely understood [19]. A more controlled study has been carried out with electrostatically-confined single localized spins in odd-electron quantum dots, where the spin-flip scattering was found to result in large TP ($\simeq 60$ μV/K) at low T [20]. The enhanced TP was attributed to spin-entropy transfer, similar to that in layered cobalt oxides [21]. Although GaAs/AlGaAs-based high mobility 2DESs form the host in many of these studies, no systematic TP experiments have been reported in unconfined 2DESs at mesoscopic length scales, which could provide crucial insight into the nature and role of intrinsic spins. Here we report experimental observation of a giant diffusion TP in delocalized (conductivity $\gg e^2/h$) 2DES in high-mobility GaAs/AlGaAs heterostructures at T < 0.3 K. We find that at most gate voltages $|S_d|$ *increases* with decreasing T down to the base lattice temperature (T_{latt}) of $\simeq 70$ mK, indicating that it cannot be explained with free noninteracting electrons in 2D. We complement thermal measurements with conventional nonequilibrium transport, and show that these results point strongly towards the existence of localized spins in 2D mesoscopic systems. Our findings may have a direct impact on the understanding of many experimentally reported, but not fully understood, phenomena in low-dimensional quantum systems, such as the '0.7-state' [14] and breakdown of Wiedemann-Franz law in quantum point contacts (QPC) [22], the zero-bias anomaly [16, 17, 18], or the anomalous Hall effect in 2DESs at ultra-low temperatures [23].

Silicon delta-doped heterostructures with thick ($\approx$ 80 nm) spacer layer of undoped AlGaAs (to minimize remote impurity scattering) were employed in our experiments. Similar devices were earlier used for nonequilib-
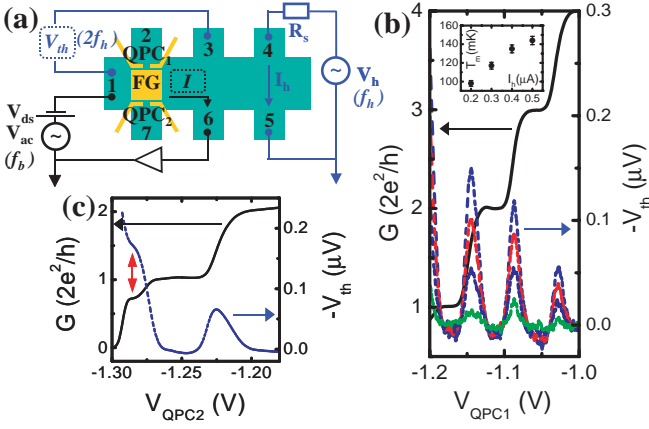
FIG. 1: (Color online) (a) Schematic of the experimental setup used to perform thermovoltage (blue lines) and conductance (black lines) measurements. (b) Characteristic peaks in V_{th} as a function of the QPC gate voltage (V_{QPC1}) for heating currents, I_h, ranging from 0.2 μA (bottom trace) 0.5 μA (topmost trace). (Inset) Electron temperature at the center of the 2D mesoscopic region obtained from the QPC TP analysis (see text). (c) A similar trace for QPC_2, where a distinct 0.7 structure was observed. Arrows mark the position of the 0.7 structure in conductance and the corresponding deviation of the thermovoltage from the semiclassical expectation. (Inset) Scanning electron micrograph of the device with a central $5\mu m \times 5\mu m$ gate used to form the mesoscopic system.

rium transport and Hall measurements [16, 17, 23]. The mobility of the electrons in these wafers were found to be in the range of $1-3 \times 10^6$ cm^2/V-s at the as-grown electron sheet density, $n_s \simeq 1 \times 10^{11}$cm^2 resulting in a long elastic mean free path ($\gtrsim 10\mu$m). A schematic of the device-structure for TP measurements with the gate assembly is shown in Fig. 1a (see inset of Fig. 1c for the SEM micrograph of the device region). Central to the design is the 5μm×5μm full gate (FG) which forms the mesoscopic device under study. A voltage (V_{FG}) on this gate tunes n_s in the 2DES directly below, thus allowing for a detailed study of the thermovoltage as a function of E_F of the mesoscopic system. The QPCs on either side of FG serve three purposes: (1) Lateral isolation of the device region from the remaining ungated 2DES, (2) validation of the measurement technique with known thermoelectric behavior of QPCs, and (3) electron temperature calibration of the mesoscopic region with respect to T_{latt} using the procedure described in Ref [24]. An oscillatory heating current ($I_h^{4,5}$) with frequency $f_h = 7.3$ Hz was used between remote leads 4 and 5. The thermovoltage detection was performed with a lock-in amplifier at $2f_h$ to ensure a purely thermal origin of the signal.

Fig. 1b shows $-V_{th}^{2,7}$ across QPC_1 as a function of the split-gate voltage V_{QPC1} for various I_h (0.2μA$\rightarrow 0.5\mu$A) with T_{latt} fixed at the base. The peaks in $|V_{th}|$ between two consecutive conductance plateaus could be scaled (not shown) for $I_h \leq 0.5\mu$A yielding the absolute electron

temperature T_m at the center of the mesoscopic region as a function of I_h (see inset of Fig. 1b). The quantitative agreement between measured TP of the QPC and the energy-derivative of its conductance (through Eq. 1) supports the independent, non-interacting electron description of the QPC at higher subbands. We also notice that, (1) the TP in our devices has a purely diffusive (and ballistic for QPC) origin, and any contribution from phonon drag is negligible, as expected for GaAs-based 2DES below ~ 300 mK [25]. (2) Scaling of thermopower for $I_h \leq 0.5\mu$A suggests thermal broadening to be negligible at these currents. (3) Fig. 1c shows the TP in QPC_2 (keeping V_{QPC1} fixed) below the first subband where a clear deviation from the Mott relation is observed near the '0.7 structure'. This deviation has been studied by Appleyard et al. [19], but its origin is yet to be completely understood. However it serves as proof that the observed TP is indeed capable of detecting signatures of many-body spin-correlated states in low-dimensional systems. For subsequent measurements of thermopower across the mesoscopic region, both QPCs were pinched off, and $V_{th}^{1,3}$ was measured after adequate amplification. The temperature difference (ΔT) across the device was estimated as $\Delta T \approx (L/\xi)[T_m(I_h) - T_{latt}]$, where $L(= 5\mu$m) and $\xi(\approx 100\mu$m) are the device length and thermal relaxation length in high-mobility GaAs/AlGaAs systems [24, 26], respectively. For electrical transport, we have used a standard ac/dc technique to measure both linear response conductivity ($G^{1,6}(V_{ds} = 0)$) and non-equilibrium differential conductivity ($dI(V_{ds})/dV$), where V_{ds} is the drain-source bias. All measurements were carried out at zero magnetic field. The electrical characteristics were recorded only at $I_h = 0$, the thermovoltages were measured at $I = 0$.

Fig. 2a shows the measured thermovoltage for our mesoscopic device at various I_h as a function of V_{FG} at $T_{latt} \simeq 70$ mK. We concentrate on the delocalized regime of the 2DES, over $G \sim (2-20) \times e^2/h$, which corresponds to $V_{FG} \gtrsim -0.52$ V, or equivalently, $n_s \gtrsim 1.8 \times 10^{10}$ cm^{-2} (see inset of Fig. 2a). A reproducible fluctuating behavior in V_{th} rides on an overall background that increases with decreasing n_s. Note that the decreasing magnitude of V_{th} with increasing V_{FG} ensures that the contribution to V_{th} from the ungated part of the 2DES between the thermovoltage leads (1 and 3) is negligible, and the measured V_{th} arises predominantly from the TP of the mesoscopic region only. The V_{th} vs. V_{FG} traces could be collapsed onto a single trace by normalizing each V_{th} with the corresponding ΔT obtained from QPC calibration. Strikingly, Fig. 2b indicates the TP of our mesoscopic device to become $\gtrsim 100\mu$V/K at low n_s, unexpectedly large for a delocalized system at subKelvin temperatures and zero magnetic field. While the qualitative agreement with the energy derivative of linear conductivity (red dotted line) provides further support to the diffusive origin of TP, the scaling also confirms that increasing I_h does not lead
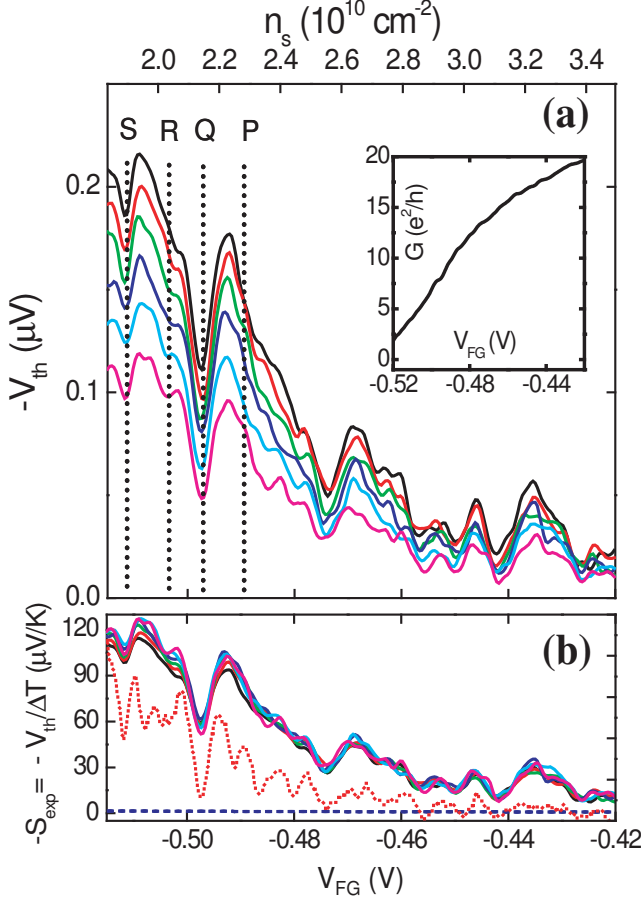
FIG. 2: (Color online) (a) Variation of $-V_{th}$ as a function of V_{FG} for I_h ranging from $0.15\mu A$ (bottom trace) to $0.25\mu A$ (topmost trace). The top axis shows the corresponding electron sheet density (n_s) of the mesoscopic system. (Inset) Equilibrium conductance (G) in this V_{FG} range. (b) Curves in (a) scaled by ΔT to obtain thermopower (S_{exp}). (Dotted) Calculated $d\ln G/dV_{FG}$ using G from Figure 2, confirming the diffusive origin of TP. (Dashed) Expected TP in the free electron picture.

to thermal broadening or substantial lattice heating for phonon drag to become important.

Increase in TP with decreasing n_s can be envisaged for free degenerate electrons, as well as for systems close to a localization transition [3, 27, 28]. The latter is plausible at $G \ll e^2/h$, when the 2DES becomes inhomogeneous deep into the band tail, and transport is dominated by classical percolation [28, 29, 30]. In our case however, $G \gg e^2/h$, and direct Hall measurements also indicate the charge distribution to be uniform [23]. Moreover, the TP never reverses its sign, ruling out sequential or co-tunneling effects in unintentional quantum dots within mesoscopic region [30, 31]. In the free electron scenario with scattering from dopant potential, Eq. 1 estimates the TP to be $\approx -\pi^2 k_B^2 T(p+1)/3|e|E_F$ ($p \approx 1.5$) [32], which is nearly two orders of magnitude lower than

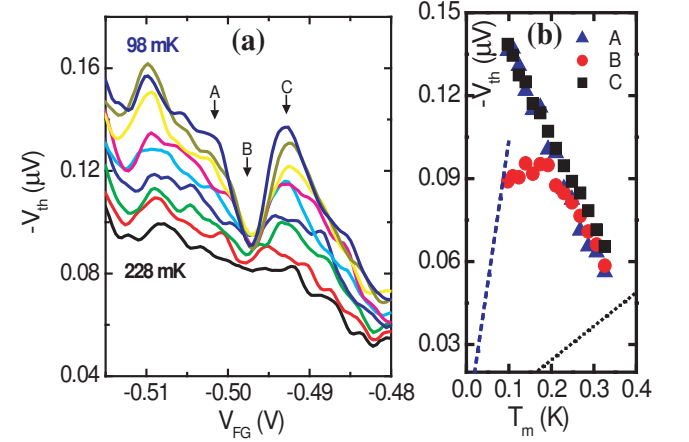the experimentally observed magnitude (dashed line in Fig. 2b).

FIG. 3: (Color online) (a) $-V_{th}$ versus V_{FG} for 98 mK $\leq T_m \leq$ 228 mK. (b) Distinct T-dependences for positions marked A, B and C in (a). Calculated T dependence expected for a free electron system (dotted) and Kondo lattice system for $T < T_K$ (dashed).

The temperature dependence of V_{th} provides further evidence of nontrivial origin of the enhanced TP in our devices. This is shown in Fig. 3a for a selected range of V_{FG} centered around a local minimum at $V_{FG} \approx -0.497$ V. Increasing T_m from 98 mK to 228 mK (at constant $I_h = 0.2$ μA), washes out strong fluctuations in V_{th}, and results in a decrease in the its overall magnitude for $T_m \gtrsim 150$ mK. Two distinct behaviors were observed (see Fig. 3b): At V_{FG}, labeled A and C, away from the local minimum, $|V_{th}|$ increases monotonically with decreasing T_m, but at the minimum, B, $|V_{th}|$ saturates at an intermediate temperature, and even decreases when T_m is reduced further. Clearly, this is not expected in the free electron scenario where $|V_{th}| \propto T_m$ (the black dotted line in Fig. 3b), neither in the proximity of localization where TP is expected to increase with decreasing T_m at all V_{FG} [3, 27, 28].

The feasibility of spontaneous spin effects in high-mobility non-magnetic 2DESs have been discussed widely [16, 18, 33], where both the background disorder and many-body exchange interaction play crucial roles. While the signature of exchange-driven Stoner ferromagnetism on the TP of a 2DES at low T is not clear, a disorder-induced two-component fluctuation in the conduction band may lead to localized spins embedded inside the delocalized Fermi sea [18], and may form a scenario similar to Kondo-lattice compounds or dilute magnetic alloys. The gate voltage V_{FG} tunes the Fermi wave-vector, and consequently the relative scales of Kondo coupling and RKKY magnetic exchange, which have characteristic signatures in the DOS in the vicinity of E_F. In nonequilibrium electrical transport, such
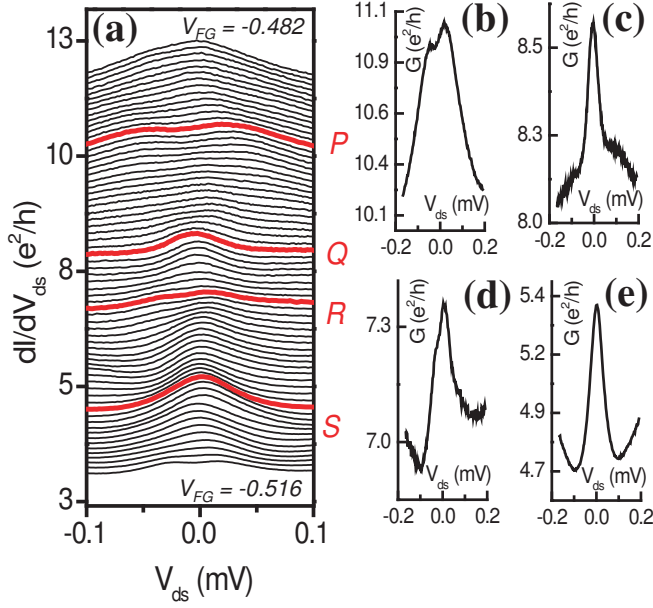
FIG. 4: (Color online) Non-equilibrium transport (a) Continuous variation of the nature of the ZBA with V_{FG}. Bold plots correspond to V_{FG} values marked P-S in Figure 2a. P,R(Q,S) show the strongest double (single) peaked ZBAs. (b)-(d) Individual ZBA corresponding to P-S in (a).

low energy structures in the DOS are manifested as a zero-bias anomaly (ZBA) in dI/V_{ds} around $V_{ds} = 0$. Fig. 4 shows two different forms of ZBA in our device as V_{FG} in changed, alternating between a single-peak at $V_{ds} = 0$ representing the Kondo resonance at individual localized spins, and a double-peaked ZBA that has a shallow minimum close to the E_F indicating finite inter-spin interaction and local magnetic ordering [18]. We note that both temperature and V_{FG}-dependence of V_{th} are directly connected to the structure of respective ZBAs. The strong minima observed in $|V_{th}|$ at -0.497 V and -0.510 V (indicated by S and Q in Figs. 2 and 4) correspond to strong single peaked ZBAs, *i.e.* when inter-spin exchange is small and Kondo-coupling dominates at low T. Analogous to heavy Fermions or dilute magnetic alloys, the T-dependence of $|V_{th}|$ in this state is nonmonotonic [10, 34](see trace B in Fig. 3), and shows a downturn just below $T_K (\approx 0.2$ K -0.3 K. (T_K was determined independently from both equilibrium and nonequilibrium transport. For details see [17]) When spin-spin exchange is strong, illustrated by traces at P and R with split ZBA, $|V_{th}|$ increases monotonically down to base temperature, implying that the giant thermopower in our devices at low temperatures could probably be a result of scattering of the conduction electrons by unscreened magnetic moments within the 2DES.

Many quantitative approaches towards thermal transport coefficients in Kondo lattice systems exist in the periodic Anderson model [34], although primarily in the context of heavy atom intermetallic compounds [5, 6, 7, 8, 11, 12]. Nevertheless, in the absence of magnetic interactions, a universal behavior of TP at $T \ll T_K$ in these systems can be expressed as $S_d = -\alpha(k_B/|e|)(T/T_K)$ [34], where $\alpha \sim \mathcal{O}[1]$. An estimate with $\alpha = 2$, and typical experimental $T_K = 0.25$ K is shown as the dashed line in Fig. 3b. Despite uncertainties in the numerics, the asymptotic behavior of trace B appears encouraging, although measurements need to be extended to lower electron temperatures for more quantitative conclusions. Two important points to note: (1) Unlike many Kondo lattice systems, or even semiconductor quantum dots [20], we do not find any change in sign of V_{th} within the experimental temperature range, remaining negative throughout. This also indicates that the average energy of the quasiparticles is negative as would be expected for a quasi-ballistic electronic system in the Kondo regime [20]. (2) We find that the modulations in V_{th} can be traced to V_{FG} as large as -0.42 V, where the large zero-bias conductance ($G(V_{ds} = 0) \approx 20 \times e^2/h$) makes the ZBA essentially undetectable. This establishes the greater sensitivity of the TP over electrical transport to detect anomalies in the local DOS near E_F.

In conclusion, we have measured unexpectedly large values of diffusion thermopower (in excess of $100 \mu V/K$) in delocalized 2D mesoscopic electron systems. Below 300 mK, the thermopower was found to increase with decreasing temperature indicating the failure of non-interacting electron model in this regime. We suggest that the observed enhancement in thermopower may be related to the formation of localized spins in the system, and draw analogies between nonmagnetic high-mobility electron devices and Kondo lattice compounds.

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